

# Unconventional Mott Transition in $K_xFe_{2-y}Se_2$

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Whether the newly discovered  $K_xFe_{2-y}Se_2$  systems are doped Mott or band insulators is key to how superconductivity emerges at lower temperature. With extant theoretical studies supporting conflicting scenarios, a more realistic approach is urgently called for. Here, we use LDA+DMFT to study this issue in  $K_xFe_{2-y}Se_2$ . We find that the undoped  $KFe_{1.6}Se_2$  system is a new kind of Mott-Kondo insulator (MKI). Electron doping this MKI drives a Mott transition to an orbital-selective non-Fermi liquid metal. Good agreement with spectral and transport responses supports our view, implying that superconductivity arises from a doped Mott insulator, as in the high- $T_c$  cuprates.

Recent finding of high- $T_c$  superconductivity (HTSC) with  $T_c \gtrsim 30$  K in  $K_xFe_{2-y}Se_2$  [1] and  $(K,Tl)Fe_xSe_2$  [2, 3] is remarkable for the following reasons: (i) While its SC  $T_c$  is comparable to that for the 1111- and 122-Fe pnictides (FePn), it occurs near antiferromagnet (AF) *insulators*: Fe composition is the tuning parameter for insulator-metal and SC instabilities. Evidence for Fe vacancy order [4] is interesting: its generic effect is to reduce LDA (local-density-approximation) bandwidths [5]. Theoretically, both Mott [5–7] and band [8] insulating states have been proposed as candidates. (ii) It intensifies the fundamental debate [9] on the degree of electronic correlations in Fe-based SC. Transport and optical data reveal insulating behavior (albeit with small gap) well above  $T_N(\delta)$ ,  $\delta$  being the electron doping. However, optical [10] and ARPES [11] studies also clearly show large-scale spectral weight transfer (SWT) (over an energy scale  $O(2.0)$  eV) as a function of temperature ( $T$ ) across the magnetic and SC instabilities, a fingerprint of Mottness. Both  $\rho(T) \simeq T$  and bad metallicity above  $T_c$  are features shared along with other non-Landau-Fermi-liquid (LFL) metals close to a Mott or selective-Mott instability [12]. ARPES [11] also seems to show a curious co-existence of Mott- and “band-insulating” spectral features in  $K_xFe_{2-y}Se_2$ . Large-scale SWT on a scale of  $O(2.0)$  eV in response to a small ( $k_B T \simeq O(10)$  meV) perturbation, however betrays the “hidden” strong correlations. Its implications for SC are intriguing: are the Mott- or “band”-like subsets of the renormalised spectra relevant for SC that emerges at lower  $T$ ? (iii) Finding of large local moment value on Fe,  $\mathbf{M}_{Fe} \simeq 3.3\mu_B$ , suggests strong electronic correlations. Even in the more *itinerant* FePn, relevance of a “dual” picture for the parent magnets is now increasingly recognised [13]: such a dual picture must be even more relevant for  $K_xFe_{2-y}Se_2$ .

These findings *necessitate* incorporation of reasonably strong multi-band electronic correlations. All five  $d$ -bands crossing the Fermi energy ( $E_F$ ) must be kept at a “minimally realistic” level in order to satisfactorily resolve the doped Mott-versus-band insulator issue above. In multi-orbital (MO) systems, sizable correlations also drive new physical effects: they induce orbital selective

(OS) bad-metallic states with no LFL coherence, naturally yielding bad metallic resistivity above  $T_c$ . An OS-Mott scenario also generically “wipes out” a subset of Fermi surface (FS) sheets from the *renormalized*, correlated band structure (Lifshitz transition). In turn, this can have far-reaching consequences for the symmetry of the SC pair function,  $\Delta(\mathbf{k})$ : details and presence or absence of gap nodes crucially depends on whether or not  $\Delta(\mathbf{k})$  intersects the *renormalized* FS of such a metal. Extant ARPES data show only electron pockets in metallic  $K_{0.8}Fe_{1.7}Se_2$  and anisotropic  $s$ -wave SC gap [14]. However, NMR  $T_1^{-1}(T) \simeq T^2$  for  $T < \frac{1}{2}T_c$  remains puzzling in this context [15]. This unsettled state of affairs calls for detailed theoretical scrutiny: it must, in view of the fundamentally conflicting views discussed above, base itself on an satisfying description of the “normal” state.

Here, we use state-of-the-art LDA-plus-dynamical mean-field theory (LDA+DMFT) [16] to address these issues. In LDA+DMFT studies of FePn, their degree of correlatedness has been the bone of contention: consensus has fluctuated between weakly [17] to sizably correlated limits [18]. In  $K_xFe_{2-y}Se_2$ , a perusal of the resistivity data [11] for non-AF but SC samples reveals that these are proximate to a Mott transition:  $\rho_{dc}(T)$  immediately above  $T_c$  is very bad metallic and quickly crosses over to an insulator-like dependence. In these cases, AF order *cannot* be responsible for insulating behavior: rather, these data show that destroying AF order reveals underlying Mottness, where (i)-(iii) above can be rationalised naturally. Clearly, electronic correlations can only get stronger as one approaches the insulator, and AF and/or orbital order can naturally arise via spin-orbital superexchange. In this work, we confirm this hypothesis, showing that a strong correlation view achieves good semiquantitative accord with a range of data in *both*, insulating and metallic phases of  $K_xFe_{2-y}Se_2$ . In particular, we clarify the co-existing Mott- and “band” insulator-like features in ARPES in a qualitatively new Mott-Hubbard scenario. Armed with these strengths, we qualitatively discuss the constraints our view imposes on mechanisms of (unconventional) SC in  $K_xFe_{2-y}Se_2$ .

We start with the experimental structure of

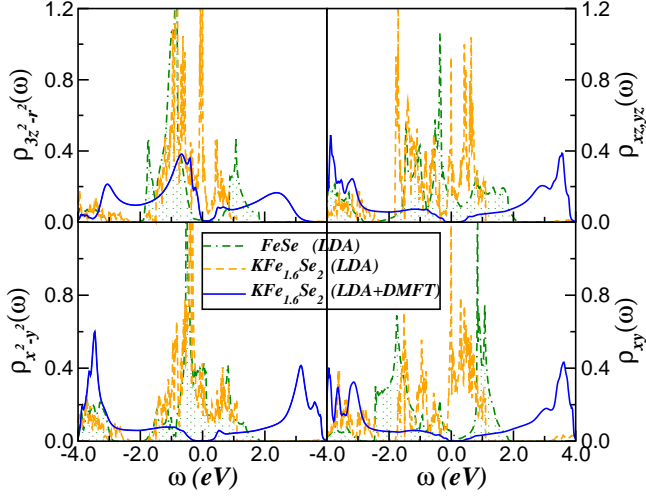


FIG. 1: Orbital-resolved LDA density-of-states (DOS) for the Fe  $d$  orbitals of FeSe (dot-dashed) and  $\text{KFe}_{1.6}\text{Se}_2$  (dashed) as well as LDA+DMFT (with  $U = 4.0$  eV,  $U' = 2.6$  eV and  $J_H = 0.7$  eV) for undoped  $\text{KFe}_{1.6}\text{Se}_2$ .

$\text{KFe}_{1.6}\text{Se}_2$  [4]. LDA calculations were performed using the linear muffin-tin orbitals (LMTO) [19] scheme, in the atomic sphere approximation: Self-consistency is reached by performing calculations with 242 irreducible  $\mathbf{k}$ -points. The radii of the atomic spheres were chosen as  $r=2.6$  (Fe),  $r=4.25$  (K) and  $r=2.7$  (Se) a.u. in order to minimize their overlap. Within LDA, the one-electron part is  $H_0 = \sum_{\mathbf{k},a,\sigma} \epsilon_a(\mathbf{k}) c_{\mathbf{k},a,\sigma}^\dagger c_{\mathbf{k},a,\sigma}$ , where  $a = xz, yz, xy, 3z^2 - r^2, x^2 - y^2$  label the (diagonalized in orbital basis) five  $3d$  bands. The MO Coulomb interactions (treated within DMFT) constitute the interaction term, which reads  $H_{\text{int}} = U \sum_{i,a} n_{i a \uparrow} n_{i a \downarrow} + U' \sum_{i,a \neq b} n_{i a} n_{i b} - J_H \sum_{i,a,b} \mathbf{S}_{i a} \cdot \mathbf{S}_{i b}$ . We use the MO iterated perturbation theory as an impurity solver for DMFT. Though not numerically *exact*, it has a proven record of recovering correct LFL metallic behavior [20] and good semiquantitative agreement in a host of real systems.

Fig. 1 shows the LDA DOS for  $\text{KFe}_{1.6}\text{Se}_2$ , whereby a clear and sizable reduction ( $\sim 20\%$ ) of the average LDA bandwidth ( $W_{\text{LDA}}$ ) relative to that for FeSe, induced by presence of Fe vacancy order, is seen. FeSe is already a bad metal close to a Mott insulator [21], and the significantly smaller  $W_{\text{LDA}}$  for  $\text{KFe}_{1.6}\text{Se}_2$  then naturally suggests emergence of a Mott insulator in the latter. Indeed, our results show a small but clear insulating gap in DMFT spectra. Several interesting features, especially germane to the above discussion, are now manifest: (i) the Mott gap is clearly orbital-dependent, i.e. intrinsically anisotropic. (ii) Examination of the orbital-resolved (imaginary parts) self-energies reveal a behavior hitherto unique to Fe-based systems. Namely,  $\text{Im}\Sigma_a(\omega)$  with

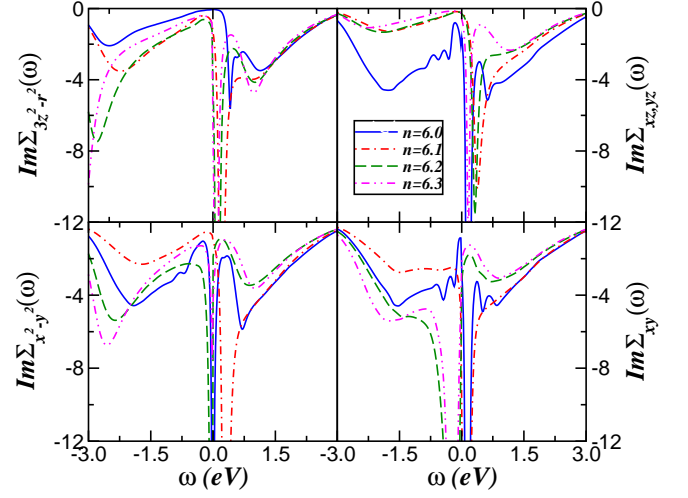


FIG. 2: Orbital-resolved (DMFT) imaginary parts of the self-energies for the Fe  $d$  orbitals of pure and electron doped  $\text{KFe}_{1.6}\text{Se}_2$  with  $U = 4.0$  eV,  $U' = 2.6$  eV and  $J_H = 0.7$  eV. The selective-Mott nature is clear.

$a = xz, yz, xy, x^2 - y^2$  clearly reveal their Mott insulating character, i.e. a pole at  $\omega = E_F (= 0)$ , as shown in Fig. 2. On the other hand,  $\text{Im}\Sigma_a(\omega)$  for  $a = 3z^2 - r^2$  simultaneously shows Kondo insulator features, i.e.  $\text{Im}\Sigma_a(\omega) = 0$  in the gap region. Thus, remarkably,  $\text{KFe}_{1.6}\text{Se}_2$  shows co-existing Mott and Kondo insulating gaps, and we dub this a Mott-Kondo insulator (MKI). One might be tempted to try to link this to the band insulator state found in LDA, especially since a Kondo insulator is an analytically continued version of a band insulator (where sizable correlations do exist above the gap scale). If electron doping would result in a metal where only the  $d_{3z^2-r^2}$  band would be driven metallic, it would necessarily mandate an *effective*, doped Kondo insulator [22], rather than doped Mott-insulator modelling. However, in ARPES data for the doped metal, the renormalised FS comprises sheets having only  $xz, yz$  orbital character, in accord with DMFT results (see below). Thus, the insulator-metal transition must now be viewed in terms of a doped Mott insulator, in accord with earlier model-based work [5, 6]. We emphasise that no such insulating state is found for  $U \leq 3.0$  eV (not shown) in absence of magnetic order.

Electron doping ( $n \equiv 6 + \delta$ , with  $\delta > 0$ ) the MKI leads to a bad-metallic state. Consistent with our results in Fig. 2, we find that only  $xz, yz$  orbitals show bad metallic behavior, characterised by absence of sharp LFL resonances at  $E_F$ , while Mott insulating behavior persists in the  $xy, x^2 - y^2, 3z^2 - r^2$  channels. Thus, we find an OS metal, and strong scattering between the Mott-localised and metallic states leads to complete suppression of the LFL quasiparticles via the Anderson orthogonality catastrophe [23], leading to emergence of anomalously broad

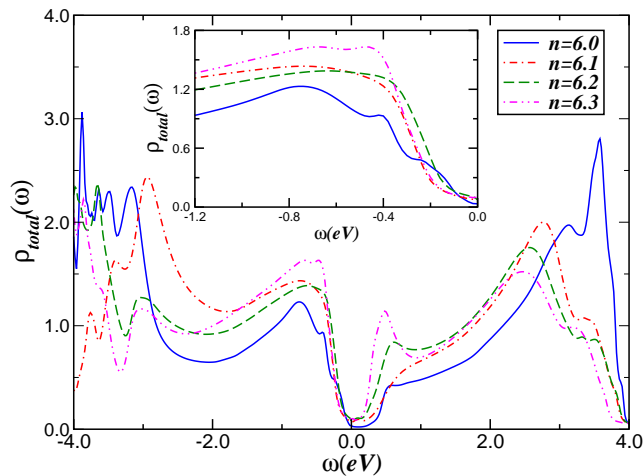


FIG. 3: Total DMFT DOS for  $\text{KFe}_{1.6}\text{Se}_2$  (main panel) as a function of electron doping  $n = 6 + \delta$ , clearly showing a low-energy pseudogap associated with bad-metallicity. Low-energy DMFT spectra (inset) in good agreement with ARPES results [2, 11, 14].

spectra in DMFT, see Fig. 3. Microscopically, infrared LFL behavior (narrow “Kondo” resonance in DMFT) is killed off by strong scattering between the Mott-localised and quasi-itinerant components of the (DMFT) matrix-spectral function, due to sizable  $U'$ ,  $J_H$ , and is intimately tied with the OS insulator-metal transition in the five-band Hubbard model we use. Thus, our selective-Mott metal is a MO counterpart of the  $\text{FL}^*$  metal [24].

If our proposal is to hold, a range of responses must find a consistent explication without additional assumptions: we now show this is indeed the case. (i) A direct comparison between DMFT spectra and (AR)PES data show very good accord: in addition to describing the overall PES lineshape very well, the DMFT spectra (inset of Fig. 3) also resolve two peaks in the low-energy ( $-0.7 < \omega < 0.0$  eV) range seen in recent work [11] for their “semiconducting” samples. Further, the renormalised FS is composed of predominantly  $xz, yz$  orbital states, and absence of hole pockets in LDA thus persists in DMFT, in accord with ARPES. Absence of LFL quasiparticles naturally implies broad ARPES lineshapes without LFL quasiparticles for electron-doped samples, as well as sizable SWT over an energy scale  $O(1.5 - 2.0)$  eV in polarised X-ray absorption (XAS) studies: this could be tested in future. (ii) Transport properties in DMFT are directly computable from the full DMFT propagators, since vertex corrections entering the Bethe-Salpeter equation are small enough to be neglected in multi-band cases [25]. In Fig. 4, we show the dc resistivity for various  $n$ . Clear (Mott) insulating behavior obtains for  $n = 6.0$ , as expected. With  $\delta > 0$ , a doping-dependent crossover from a “high”- $T$  insulator to a low- $T$  (lower inset in Fig. 4) bad metal obtains. Ob-

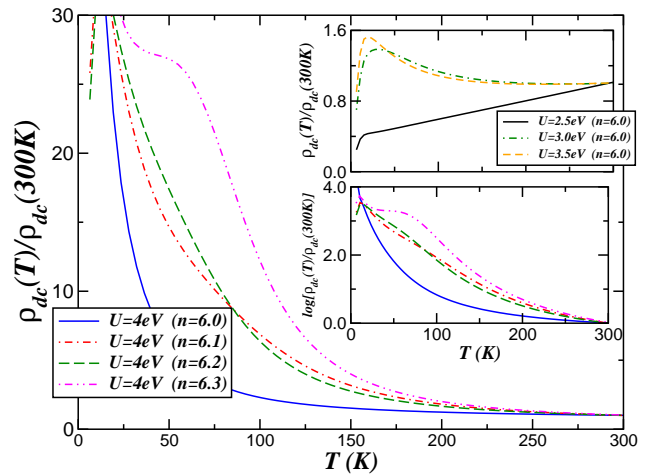


FIG. 4: Resistivities for insulating (solid curve) and electron-doped phases of  $\text{K}_x\text{Fe}_{2-y}\text{Se}_2$  as a function of  $T$  in good accord with transport data [3, 26].

viously, this crossover scale, marked by the maximum of  $\rho_{dc}(T)$ , increases with  $\delta$ . The upper inset of Fig. 4 shows that small  $U$  yields metallic behavior up to high  $T$ , in conflict with data. At the very least,  $U > 3.5$  eV is needed to obtain accord for  $\delta = 0$ , but correct insulating behavior is only found for  $U \cong 4.0$  eV. (iii) Further corroborative support comes from optical conductivity. In Fig. 5, we show the total optical conductivity,  $\sigma(\omega)$ , within DMFT, using the multi-band version of the general DMFT formalism [25].  $\sigma(\omega)$  rises with  $\omega$  at small  $\omega$ , and the optical gap,  $\Delta \simeq 0.3$  eV, is in good accord with data [10]. Further, as also seen, bad-metallic conductivity, along with large-scale ( $> O(2.0)$  eV) transfer of dynamical spectral weight, obtains for  $\delta > 0$ . It is particularly noteworthy that good accord is seen not only at low energy ( $0 < \omega < 0.3$  eV), but up to 1.0 eV, as seen by direct comparison with data (inset to Fig. 5). Comprehensive accord with PES and (dc and ac) conductivities thus provides strong support to the view that  $\text{KFe}_{1.6}\text{Se}_2$  is a Mott insulator (MI), and that the doping-driven transition to a bad metal must then be viewed in terms of a doped MI.

These findings put strong constraints on mechanisms of SC. Since LFL quasiparticles are not stable excitations in the bad metal, instabilities to ordered states via (particle-particle) BCS-like pairing of well defined LFL quasiparticles are untenable. In non-LFL, bad metals coherent one-electron hopping term is irrelevant in an RG sense [23]. Hence, ordered states can only arise via *coherent* two-particle hopping, which becomes more relevant when one-electron hopping term is irrelevant: the situation is analogous to coupled  $D = 1$  Luttinger liquids [27]. Given local incoherent metallic state(s) within DMFT, the above analogy tells us that resi-

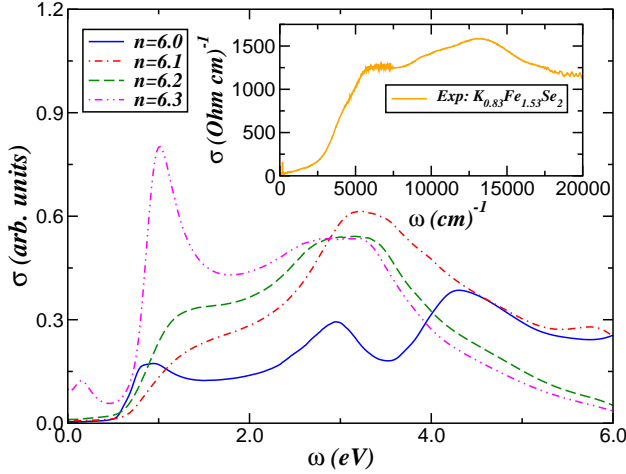


FIG. 5: (Color online) Optical conductivity for insulating (solid curve) and electron-doped phases of  $K_xFe_{2-y}Se_2$  as a function of energy. The good accord with experimental results (inset) [10] up to  $\Omega \simeq O(1.0)$  eV is seen.

ual, inter-site and inter-orbital (in multi-band systems) two-particle interactions can generate ordered states directly from the bad metal. This is the philosophy used earlier [28] for the 1111-FePn systems. As in earlier work, restricting ourselves to the  $xz, yz$  orbital sector, the effective pair-hopping term (second order in  $t_{ab}$ ) is  $H^{(2)} \simeq -1/2 \sum_{k,k',a,b} V_{ab}(k,k') c_{ka\sigma}^\dagger c_{b,k',\sigma'}^\dagger c_{b,k',\sigma'} c_{ka\sigma}$ . Decoupling  $H^{(2)}$  in the particle-hole and particle-particle channels gives  $H_{MF}^{(2)} = \sum_{k,a,b} [(\Delta_{ab}^{(1)}(k) c_{ka\sigma}^\dagger c_{kb\sigma} + h.c.) + (\Delta_{ab}^{(2)}(k) c_{ka\uparrow}^\dagger c_{kb\downarrow}^\dagger + h.c.)]$ , where  $\Delta_{ab}^{(1)}(k) = \langle \gamma(k) c_{kb\sigma}^\dagger c_{ka\sigma} \rangle$  and  $\Delta_{ab}^{(2)}(k) = \langle \gamma(k) c_{kb\sigma} c_{ka-\sigma} \rangle$  with  $\gamma(k) = \cos k_x + \cos k_y + \alpha \cos k_x \cos k_y$  for the frustrated case of Fe-based systems. These represent orbital nematic (with orbital order and lattice distortion) [29] and inter-orbital pairing [28] instabilities. Extending DMFT to study both these orders in  $K_xFe_{2-y}Se_2$  is more problematic, however: the large moment,  $\mu_{Fe} = 3.3\mu_B$ , the block-spin moment  $M \simeq 11\mu_B$ , and the block spin-AF order [30] suggests that both, possible orbital order [31] with lattice distortion and subsequent AF, as well as SC instabilities must involve coupling between four-site plaquettes, beyond what our  $H_{MF}^{(2)}$  would give. Since the Mott transition already occurs at high  $T$ , a way to proceed might involve using the present DMFT results as a template for deriving an appropriate low-energy, plaquette-centered model using the *active*  $xz, yz$  orbital states to address these issues as recognised by Baskaran in a different approach [32]. This is currently underway, and will be reported in future.

In conclusion, using LDA+DMFT for a minimally realistic five-band Hubbard model, we resolve the issue of

a doped Mott- vs band insulator physics in  $K_xFe_{2-y}Se_2$  systems in favor of the former, Mott view. Good quantitative accord with key spectral and transport data in a sizably correlated picture confirms this view, and strongly suggests close underlying similarities (in spite of very different chemistry) between SC emerging here from a doped multi-orbital Mott-Kondo-insulating state with  $d$ -wave SC in doped high- $T_c$  cuprates.

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